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Unusual Optical Properties of Aligned Carbon Nanotube Mats in Infrared Energy Region

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ABSTRACT: We studied the optical properties of aligned carbon nanotube mats for photonic device applications. We employed ab-initio density functional calculations in the linear combination of atomic orbital formalism. The calculated dielectric functions of the semiconducting carbon nanotube mats show a strong anisotropy when the electric field of the light is parallel or perpendicular to the tube axes. Especially, there are strong optical absorptions near the band edges in the infrared energy region, when the electric field of the light is parallel to the carbon nanotube axes. The unusual optical properties of the semiconducting carbon nanotube mats present a new opportunity for applications in electro-optical devices in the infrared energy region.

Keywords: carbon nanotube mats; optical properties; density functional calculation.

1. Introduction

Carbon nanotubes possess unique electronic and optical properties that are very useful for building novel electro-optical devices at nanometer scales. Recently, Fagan et al. reported measurements of the full intrinsic optical anisotropy of isolated single-wall carbon nanotubes (SWNTs). They stretched the DNA-wrapped SWNTs in polyacrylic acid (PAA) to make transparent films of aligned SWNTs. By combing absorption spectroscopy with transmission ellipsometry and polarization-dependent resonant Raman scattering, they obtained the real and imaginary parts of the SWNT permittivity (or dielectric function) from aligned semiconducting SWNTs.[1] Although the samples included multiple SWNT chiralities, they observed strong anisotropy in the real $\varepsilon_v^{'}$ and imaginary parts $\varepsilon_v^{'''}$ of permittivity in the absorption spectra for polarization in parallel or normal to the SWNT symmetry axis. Previously, Wu et al. reported ultrathin, transparent, optically homogeneous, electrically conducting films of pure single-walled carbon nanotubes.[2] The films exhibit optical transmittance comparable to that of commercial indium tin oxide in the visible spectrum, but far superior transmittance in the technologically relevant 2- to 5-micrometer infrared spectral band. Kim et al. also observed highly polarized absorption and photoluminescence of stretch-aligned single-walled carbon nanotubes dispersed in gelatin films.[3] Li et al. reported polarized optical absorption spectra of single-walled 4 Å carbon nanotubes arrayed in the channels of an AlPO4-5 single crystal.[4] The measured absorption spectra agreed well with ab-initio calculations based on the local density functional approximation. Machon et al performed ab-initio density functional calculations for the optical properties of 4 Å-diameter single-walled carbon nanotubes.[5] The calculated optical properties confirmed the experimental results of Li et al. for the strong anisotropy of the optical response of carbon nanotubes.

We performed ab-initio density functional calculations and studied the optical properties of aligned SWNTs that form carbon nanotube mats (CNTMs) or films. We are especially interested in the optical properties of CNTMs that were constructed from semiconducting SWNTs for optoelectronic applications. Although the fabrication of aligned carbon nanotube mats with the same chirality (or diameter) of SWNTs has not been achieved yet, we present the unusual optical properties of the aligned carbon nanotube mats in infrared energy region, aiming to stimulate further research for photonic applications.

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14. ABSTRACT
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2. Method

We performed ab-initio quantum calculations that are based on the density functional theory of Hohenberg-Kohn and Kohn-Sham.[6,7,8,9,10] We utilized the liner combination of atomic orbitals (LCAO) method in solving the Kohn-Sham equations. We employed an extended basis set that includes atomic orbitals of C(1s2s3s 2p3p). Here C(3s 3p) are the extra orbitals that are used to augment the basis set to account for possible charge diffusion and polarization in the CNTMs. As identified in the experimental studies, single-walled carbon nanotubes are self-assembled in a triangular lattice in bundles, strands, or mats.[11],[12] We constructed the carbon nanotube mats by aligning the SWNTs in an array of the triangular lattice. We vary the distances between the SWNTs to simulate the different samples of the interactions in the mats. We utilized ab-initio density functional calculations to study the electronic structure of the aligned carbon nanotube mats. For the studies of the optical properties of the aligned CNTMs for photonic applications, we are particularly interested in the semiconducting SWNTs. We utilized the calculated electronic energy levels and related wave functions to calculate the real and imaginary parts of the dielectric function of CNTMs as a function of photon energy.

3. Results

We first constructed a carbon nanotube mat utilizing SWNT (16, 0) as the basic building block. The diameter of individual SWNT (16, 0) is 1.25 nm, which is comparable to the ones that were experimentally studied by S. Kazaoui et al.[13] There are 64 atoms in the tube unit cell of SWNT (16, 0). The large number of atoms per unit cell that enter into ab-initio calculations presents some technical challenges. We utilized a real space approach of LCAO method in the calculations. Different from the work of Guo et al,[14] we aligned the SWNTs (16, 0) in a triangular lattice to form the mat. Guo et al used a supercell geometry and the nanotubes were aligned in a square array with the closest distance between the adjacent nanotubes being at least 6 Å. In this work, we performed ab-initio total energy calculations for the carbon nanotube mats at various tube-tube distances. The total energy minimization found the stable tube-tube distance at 3.5 Å, which is close to the interplanar distance (3.35 Å) of graphite.

The calculated dielectric function revealed a strong anisotropy when the electric field (E) of the light is parallel or perpendicular to the tube axis. Fig. 1 presents the calculated dielectric function of the aligned nanotube mat, for the tube-tube distance of 3.5 Å between the individual SWNTs (16, 0). When the electric field of the light is polarized in parallel to the tube axis, the imaginary part of the dielectric function ε_2 has a strong peak at the photon energy of about 0.58 eV, as in Fig. 1(a). The height of the first peak of ε_2 in Fig. 1(a) can reach a value of about 60, which is about a factor of seven higher than that of the first peak of ε_2 (at 0.9 eV) in Fig. 1(b). The strong peak in the imaginary part of the dielectric function (ε_2) in the infrared energy region in Fig. 1(a) shows a strong anisotropy in the optical properties of CNTM and is spectacularly different from that of conventional optical materials. The above strong peak in ε_2 indicates that the carbon nanotube mats can be used as novel photonic sensors in the infrared region. Such unusual optical properties in semiconducting carbon nanotube mats present a new opportunity for applications in new electro-optical devices.

We then constructed another CNTM by arranging individual SWNTs (10, 0) in a triangular lattice. The tube-tube distance is 3.34 Å. The diameter of individual SWNT (10, 0) is 7.83 Å. Fig. 2 presents the calculated real and imaginary part of the dielectric function of the carbon nanotube (10, 0) mat. The general feature of the dielectric function of the aligned carbon nanotube (10, 0) mat is similar to that of the (16, 0) mat. The calculated imaginary part of the dielectric function in Fig. 2(a), for the electric field of the light parallel to the tube axis, shows a strong peak near the absorption edge at about 0.75 eV. When the electric field of the light is perpendicular to the tube axis, the absorption is much weaker, as shown in Fig. 2(b). This feature again presents a strong anisotropy in the optical properties of the aligned carbon nanotube mat.

We further constructed another CNTM by arranging SWNTs (8, 4) in a triangular lattice. The tube-tube distance in the mat in the triangular lattice is 3.34 Å, as that in the carbon nanotube (10, 0) mat. The diameter of individual SWNT (8, 4) is 8.29 Å. We performed ab-initio density functional computations to study the optical properties of the carbon nanotube (8, 4) mat. Fig. 3 presents the calculated optical properties, i.e. the real and imaginary parts of the dielectric functions as functions of the photon energy. Fig. 3 (a) and (b) present the results for the electric field of the light parallel and perpendicular to the tube axis, respectively. Similar to the carbon nanotube (16, 0) and (10, 0) mats, there is again a strong peak in the imaginary part ε_2 of the dielectric function near the absorption edge at about 0.89 eV, when the electric field of the light is parallel to the tube axis. The imaginary part (ε_2) of the dielectric function can reach a value of about 49 at the photon energy of 0.89 eV. There is a strong anisotropy in the optical properties in the carbon nanotube (8, 4) mat, as shown in Fig. 3 (a) and (b).

4. Conclusion

In this work, we studied the optical properties of the aligned carbon nanotube (16, 0), (10, 0) and (8, 4) mats for photonic device applications. The calculated dielectric function of the aligned carbon nanotube mats present a strong anisotropy when the electric field of the light is polarized in parallel or perpendicular to the tube axes. Especially, there are strong peaks in the imaginary part of the dielectric function near the absorption edges, when the electric field of the light is polarized in parallel to the carbon nanotube axes. The unusual optical properties of the semiconducting carbon nanotube mats present a new opportunity for applications in new electro-optical devices in the infrared energy region.

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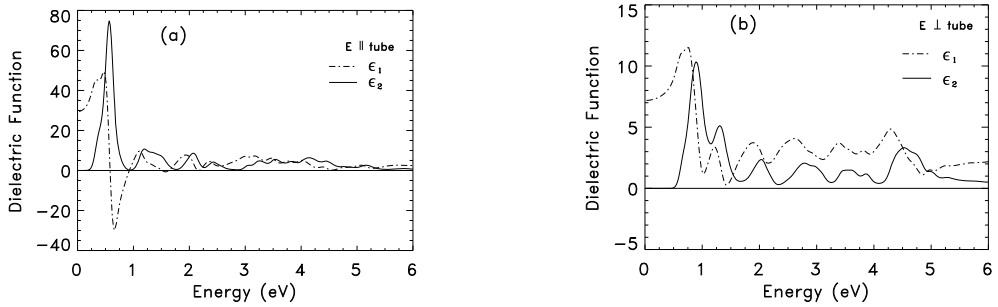

Fig. 1. The calculated real and imaginary parts of the dielectric function, $\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$, of the carbon nanotube (16, 0) mat which has the tube-tube distance of 3.5 Å between the SWNTs (16, 0).

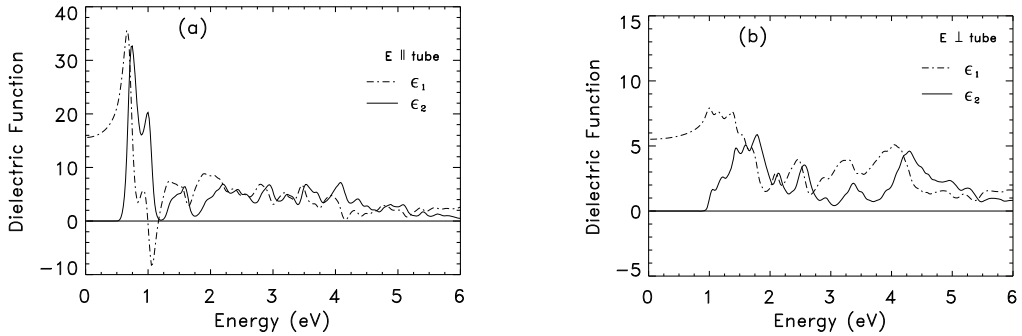

Fig. 2. The calculated real and imaginary parts of the dielectric function of the carbon nanotube (10, 0) mat which has the tube-tube distance of 3.34 Å between the individual SWNTs (10, 0).

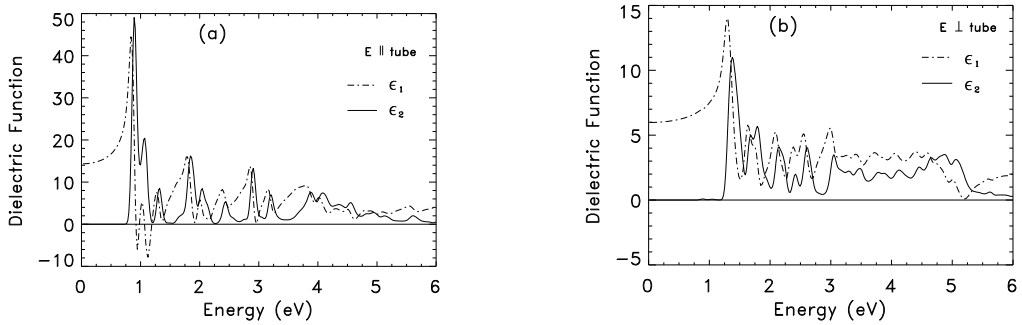

Fig. 3. The calculated real and imaginary parts of the dielectric function of the carbon nanotube (8, 4) mat which has the tube-tube distance of 3.34 Å between the chiral SWNTs (8, 4).